

Correction of Non-Linearity in Detectors for Electron Spectroscopy

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INTRODUCTION

The intensity levels reached in many third-generation synchrotron radiation experiments on solids have been found to exceed the linear response range of the final detection system involved. In electron spectroscopy, Seah and co-workers have previously discussed methods for detecting such non-linearity, including measurements with laboratory x-ray sources [1]. As a particular case involving synchrotron radiation, non-linearity has been noticed by several groups in using the Gamdata/Scienta electron spectrometers, with this behavior extending even to fairly low countrates [2-8]. For example, prior work on multi-atom resonant photoemission (MARPE) by several groups was strongly affected by this particular non-linearity [3-6]. While data related to the MARPE effect have been corrected for such non-linearities in prior publications [6b,7,8], we have found that other measurements such as the quantitative analysis of complex oxides via core-level intensities can be strongly influenced by this non-linearity, even when the exciting energy is far away from any resonance [9]. Similarly, measurements of relative intensities in angle-resolved valence spectra can also be significantly altered [10]. It is thus of interest to develop accurate and broadly utilizable procedures for correcting for non-linearity with any detection system. Although we will use one spectrometer system as an example, the methodologies discussed here should be useful for many other cases.

CORRECTION METHODOLOGIES AND EXPERIMENTAL RESULTS

In Figure 1(a), we show broad-range survey spectra from a Cu(110) sample, as excited by Al K α radiation with various power levels and detected by a Scienta SES200 spectrometer and its standard microchannel-plate/phosphor/CCD detection system. Since the high voltage was held constant, this is verified experimentally to lead to x-ray flux being proportional to power [7]. The various spectra have been normalized to one another at the lowest-countrates, just above the valence region. If the detector were linear, all spectra should lie on top of one another, but it is evident that they are not, with factors of up to 4x separating them in the higher intensity regions at lower kinetic energy.

We now consider two methods for correcting for non-linearity in such spectra:

1. Measurement of flat-background reference intensity as a function of incident x-ray flux: In the most obvious method, a flat background region in a spectrum from a sample with a stable surface can simply be measured as a function of incident x-ray flux, with non-linearity then being reflected in any deviation of a plot of measured intensity vs. x-ray flux from a straight line [7]. One might term this a "partial yield" measurement of detector response. This curve can then be fit to a convenient polynomial function such that any non-linearity can be described finally via:

$$I_m(I_t, I_x) = b_0 + b_1 I_t(I_x) + \sum_{n=2}^{n_{\max}} b_n I_t^n(I_x), \quad (1)$$

where I_m = the measured countrate, I_t = the true countrate, I_x = the incident x-ray flux, the b_n are empirical expansion coefficients, and n_{\max} is some maximum order chosen to adequately fit the data (roughly 5 in one prior analysis [7]). The coefficient b_0 thus represents the dark current in the absence of any excitation, and this will often be negligible, or at least can be simply subtracted from all measurements. The overall counting efficiency ε is now defined simply as

$$\varepsilon(I_m) = \frac{I_m(I_t, I_x) - b_0}{I_t(I_x)} = b_1 + \sum_{n=2}^{n_{\max}} b_n I_t^{n-1}(I_x). \quad (2)$$

Actual measured spectra can now be generally written as vectors $[I_{mi}(E_i)]$ over different energies E_i , and can be corrected to yield true spectra $[I_{ti}(E_i)]$ simply by dividing by ε , or

$$I_{ti}(E_i) = [\varepsilon(I_{mi})]^{-1} I_{mi}(E_i) \equiv \delta(I_{mi}) I_{mi}(E_i), \quad (3)$$

where the quantity $\delta(I_{mi})$ is defined by this equation.

Some measured vs. true countrates derived in this way are shown in Figure 1(c), and at minimum a significant quadratic correction term is evident. This procedure has been used in a prior analysis of detector non-linearity by Kay and co-workers [7,8], and it is found to yield excellent correction for such effects in the Scienta system, including the MARPE data discussed previously.

2. Analysis of broad-scan survey spectra at different incident x-ray fluxes:

Consider a set of N survey spectra $I_{mi}^j(I_x^j, E_i)$ measured on the same sample, with incident fluxes $j = 1, 2, \dots, N$, as illustrated shown in Figure 1(a). The intensities here span a range of approximately 40x, thus sampling the detector response very fully. We further assume that the true countrate I_{ti}^j (minus dark current as needed) for a given flux I_x^j , and energy E_i , can be expressed via a simple proportionality to flux, and that the true spectra can also be described by another power series in the measured spectra, as

$$I_{ti}^j(I_x^j, E_i) = I_x^j I_0(E_i) = \sum_{k=1}^P a_i I_{mi}^k(E_i), \quad (4)$$

where $I_0(E_i)$ = the true spectral shape in energy, $j = 1, 2, \dots, N$; $i = 1, 2, \dots, M$ = no. of channels in energy, and P is the maximum order of the polynomial needed to adequately describe the data (roughly 9 as a conservative number). Further assuming that the detector is linear at very low count rates such that, and then substituting for I_{ti}^j and rearranging, permits expressing the data for all N spectra as:

$$\underbrace{-\left[\frac{I_{mi}^1(I_x^1, E_i)}{I_x^1} - \frac{I_{mi}^j(I_x^j, E_i)}{I_x^j} \right]}_{=\mathbf{B}} = \sum_{k=2}^P \underbrace{\left[\frac{I_{mi}^k(I_x^1, E_i)}{I_x^1} - \frac{I_{mi}^k(I_x^j, E_i)}{I_x^j} \right]}_{=\mathbf{M}} = \underbrace{a_i}_{=\mathbf{A}}. \quad (5)$$

In matrix form, \mathbf{B} is an $(N-1) \cdot M$ long column vector, \mathbf{M} an $(N-1) \cdot M$ by $(P-1)$ matrix, and \mathbf{A} a $(P-1)$ long column vector. Eq. (5) is an over-determined system of linear equations and can be solved for the maximum likelihood a_i 's in \mathbf{A} by finding the minimum of $|\mathbf{B} - \mathbf{MA}|^2$, i.e. by solving for $0 = \nabla_{\mathbf{A}} |\mathbf{B} - \mathbf{MA}|^2 = 2\mathbf{M}^T \mathbf{M} - 2\mathbf{M}^T \mathbf{B}$ (superscript \mathbf{T} = transpose). The polynomial coefficients in can be obtained by a standard "LU" decomposition [11] or simply by matrix inversion:

$$\mathbf{A} = (\mathbf{M}^T \mathbf{M})^{-1} \mathbf{M}^T \mathbf{B}. \quad (6)$$

where $(\mathbf{M}^T\mathbf{M})$ is a small $(P-1)$ by $(P-1)$ matrix. For better numerical precision in the matrix inversion, the measured counts should vary from 0 to 1, which can be achieved by trivial normalization.

Some results obtained with both methods are shown in Figure 1(d), where the true counts are plotted as a function of measured counts, and essentially identical results for the curve are found from both methods. Finally, in Figure 1(b), we show the same spectra as Figure 1(a), but with the correction applied: it is clear that all normalized spectra for different fluxes coincide to a high accuracy, thus permitting quantitative spectroscopy to be performed with this detector.

CONCLUSIONS

In summary, we have presented two distinct methods for correcting for non-linearity in detection systems, and demonstrated their validity for the specific case of the Scienta SES 200 spectrometer. Applications to this and other spectrometer systems for electrons and soft x-rays should be possible.

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Figure 1. (a) Survey spectra from a Cu(110) sample, obtained with AlK α excitation at various x-ray powers and constant voltage, normalized to be equal at the lowest countrates near zero binding energy. (b) Survey spectra for the four lowest power settings after non-linearity correction. (c) Plot of true countrate vs. measured countrate, as derived from Method 1. Countrates expressed in Hz per pixel in the CCD camera [7,8]. (d) Plot of measured countrate vs. true countrate as derived from both Methods 1 and 2.

